Atmospheric Measurements During *Polarstern* Cruise ANT VII/1, 54° N to 32° S: An Overview

U. PLATT[1,*], J. RUDOLPH[1], T. BRAUERS[1], and G. W. HARRIS[2]

Abstract. A comprehensive set of trace gas concentrations and meteorological parameters were measured simultaneously during a cruise of the research vessel *Polarstern* from Bremerhaven (54° N, 8° E) to Rio Grande (32° S, 52° W) during the period from 15 September to 9 October 1988. This paper describes the general features of the cruise and summarizes the measurements made, the techniques employed, and the placement of the instruments on board the ship. The synoptic data base is used to characterize the nature and possible origins of the air masses encountered during the cruise and to draw some general conclusions from the measurements.

Key words: Maritime troposphere, trace gas measurements, oxidant formation.

1. Introduction

Sources, sinks, and transformation processes in marine air can contribute significantly to the global budgets of many trace species, such as ozone (Logan, 1985), hydrocarbons (Rudolph, 1988), and reduced sulphur compounds (Raemdonck and Andreae, 1983). Although more than two-thirds of our planet is covered by oceans, much remains to be discovered about concentrations and transformation mechanisms of trace gases in the oceanic atmosphere: for instance, it is not clear whether the remote marine troposphere is a nett source or sink of ozone (Liu *et al.*, 1980).

Since most of the trace gas cycles are chemically coupled, it is of particular importance to measure a comprehensive set of species simultaneously. Such measurement sets not only enhance our knowledge about the distribution of man-made and natural species, but can also be used to test model simulations of chemical interactions in the undisturbed atmosphere. In particular, it is possible to obtain a better quantification of the oxidizing capacity of the atmosphere.

Consequently among the scientific goals of the 1988 *Polarstern* atmospheric chemistry campaign were: the investigation of the distribution and concentration levels of atmospheric oxidants such as ozone or hydrogen peroxide; the assessment of the influence of long-range transport on the trace gas composition of the marine boundary layer; the study of the contribution of air-sea exchange to the budgets of hydrocarbons and sulphur species and the determination of the radiation budget and photolysis frequencies of ozone and NO_2 in the tropics.

The research vessel *Polarstern* started its cruise at Bremerhaven, Germany (54° N, 8° E) on 15 September 1988, and the leg of the journey described here ended at Rio Grande, Brazil (32° S, 52° W) on 9 October. To allow a meridional cross-section, the cruise followed the 30° W meridian from (30° N, 30° W) to (30° S, 30° W), see Figure 1. A total of 15 research groups participated in the atmospheric chemistry program on this cruise. It is the purpose of this paper to give a synopsis of the measurements and techniques employed, to present 'basic' measurements (like meteorological parameters) and, based on general features of the measurements, to characterize the air masses encountered during the cruise. These data form the background for the detailed interpretation of the individual measurement sets reported separately (see Table I).

The citations in Table I refer to the proceedings of the conference on the 'Physico-chemical Behaviour of Atmospheric Pollutants', held at Varese, Italy in September 1989, where most of the data from the *Polarstern* cruise was reported in preliminary form. Papers on the majority of the data sets and their interpretation appear in this issue of the *Journal of Atmospheric Chemistry* and are referred to in the text.

2. The Placement of the Instruments on Board the *Polarstern*

Obviously a Diesel powered vessel represents a strong local source of pollutants such as NO_x, hydrocarbons, or CO, and, inevitably, the trace gas measurements had to be made a short distance from this source. Therefore, in order to minimize possible contamination from the ship, all instruments measuring atmospheric parameters were mounted as high as possible above the sea surface and forward of the engine exhaust. Figure 2 shows the siting of the various experiments. Three placements were available, the crow's nest (A, 27.5 m above sea level), the compass deck (B, 21 m above sea level), where three containers were mounted to house most of the instruments, and the observation room one deck below the compass deck (C, 17 m above sea level). Across the full width of the compass deck (site B), a 20 m base path multi-reflection cell for DOAS measurements was mounted (Platt *et al.*, 1990). Table I lists the site for each individual experiment.

At sites A and B contamination-free sampling would be possible when the relative wind direction (counting bow as zero) was between 270° and 360° or 0° and 90°, provided the relative wind speed was sufficiently high, ($<\sim 2$ m s^{-1}; since the ship's speed was ~ 5 m s^{-1} this condition always obtained when the true wind was from the southern quadrants). For site C, the conditions are not quite so favourable and there was a higher contamination risk based on the relative wind speeds and directions. Few contamination problems were encountered, but during sections of the trip certain experiments were or may have been affected by the ship's emissions (Rohrer and Brüning; Harris *et al.*, 1992 – this issue).

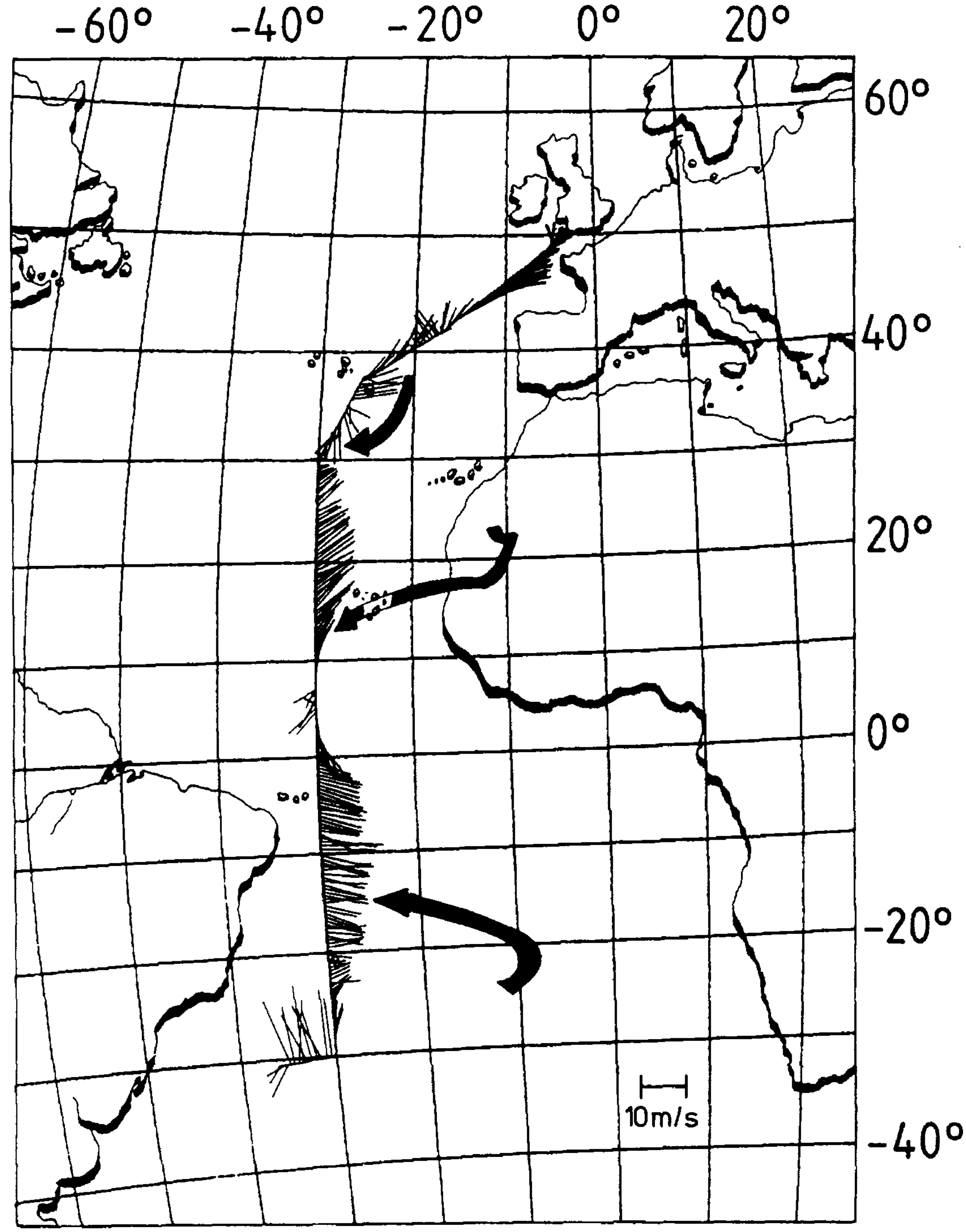

Fig. 1. Chart of the *Polarstern* cruise ANT VII/1, lines at course indicate true wind direction and speed (see scale at lower right). Arrows indicate calculated sample three-day airmass trajectories.

3. Meteorology and Cruise Track

Meteorological parameters (air and water temperatures, air pressure, relative humidity) were measured using standard meteorological instrumentation and

Table I. Atmospheric trace species measured during ANT VII/1

Parameter	Technique	Site	Reference
Oxidized Nitrogen Species:			
NO_y, NO, NO_2	CL (gas-phase)	C	Brüning, Rohrer (1990)
NO_2	TDLS	B	Harris *et al.* (1990)
HNO_3	LPFF	C	Papenbrock *et al.* (1990)
HNO_3	Filter sampling	A	Papenbrock *et al.* (1990)
PAN	GC	B	Müller *et al.* (1990)
NO_3, NO_2	DOAS-LPC	B	Platt *et al.* (1990)
Oxidants:			
O_3 (sea level)	UV	B	Callies *et al.* (1990)
O_3 (profile)	Electrochemical Balloon-sonde	D	Smit *et al.* (1990)
H_2O_2	CL (liquid-phase)	B	Jacob + Klockow (1990)
H_2O_2	TDLS	B	Harris *et al.* (1990)
Halogen Species:			
IO	DOAS-LPC	B	Platt *et al.* (1990)
HCl	TDLS	B	Harris *et al.* (1990)
Carbon Species:			
NMHC	GC	B	Koppmann *et al.* (1990)
CO	GC	B	Koppmann *et al.* (1990)
CO	TDLS	B	Harris *et al.* (1990)
Aldehydes	Derivatisation	B	Harris *et al.* (1990)
	HPLC	A	Carlier *et al.* (1990)
HCHO	TDLAS	B	Harris *et al.* (1990)
$^{14}CH_4$	AMS	C	Levin (unpublished results)
Sulphur Species:			
MSA	GC	B	Bürgermeister *et al.* (1990)
DMS, CS_2, COS	GC	B	Staubes *et al.* (1990)
DMS	GC	A	Pashalidis *et al.* (1990)
Aerosol Properties:			
Nr. density	CNC	B	Callies *et al.* (1990)
Area ($r > 0.3$)	Nephelometer	B	Callies *et al.* (1990)
Elemental composition	*n*-activation	A	Losno *et al.* (1990)
NO_3^-	Filter sampler	A	Papenbrock *et al.* (1990)
Rain and Seawater:			
MSA	IC	B	Bürgermeister *et al.* (1990)
H_2O_2	CL (liquid-phase)	B	Jacob + Klockow (1990)
NMHC	GC		Plass *et al.* (1990)
Meteorol. Parameters, Radiation:			
Wind speed	standard meteorol.	A	(this paper)
Wind direction	instrumentation	A	
Global radiation		B	
$J(O_3)$	photoelectric-	B	Callies *et al.* (1990)
$J(NO_2)$	sensors	B	Callies *et al.* (1990)
UV-B		A	Behr (1990)

Measurement Techniques:
TDLS: tunable diode laser spectroscopy
LPFF: laser-photolysis fragment-fluorescence
GC: gas chromatography
CL: chemiluminescence
IC: ion chromatography
DOAS: differential optical absorption spectroscopy
LPC: Long path cell
HPLC: high performance liquid-phase chromatography
AMS: accelerator mass spectroscopy
CNC: condensation nuclei counter

Instrument Sites on the Polarstern (see also Figure 2):
A: mast
B: compass deck
C: observation room
D: balloon sonde

recorded by the integrating data system (INDAS) of the *Polarstern*. Figure 3 shows the measured wind speed and direction relative to the vessel as well as the 'true' values obtained by correcting for the movement of the vessel. Air and water temperature as well as relative humidity (RH) data are shown in Figure 4. The temperature of both the air and sea showed a parallel latitudinal trend, with maximum values occurring near 10° N. In the latitude range from 30° N to 25° S the RH stayed in the range 60 to 85%, with the exception of in the Intertropical Convergence Zone (ITCZ), where the humidity was close to 100%.

The true wind directions and speeds encountered are shown in Figure 1. During the first leg (Bremerhaven to 30° N, 30° W) varying directions were encountered. From 30° N, 30° W to near the ITCZ (10° N, 30° W), the course crossed the NE trades region. In the ITCZ region (10° N, 30° W to 4° N, 30° W) again varying wind direction prevailed. The SE trades region extended from 4° N, 30° W to 25° S, 30° W. South of this point, the weather was influenced by a strong low pressure system centred near 40° S, 47° W resulting in northerly winds at the ship.

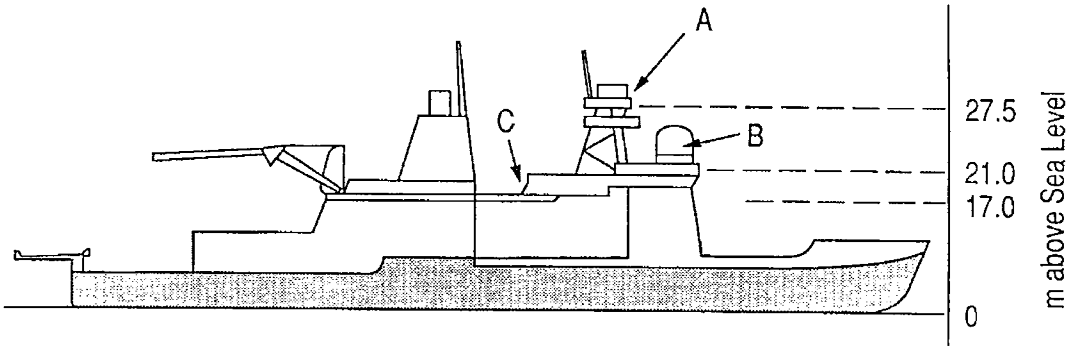

Fig. 2. Location of the various instruments (i.e. inlet tubes) on board the Polarstern. A the crow's nest, B compass deck, C observation room.

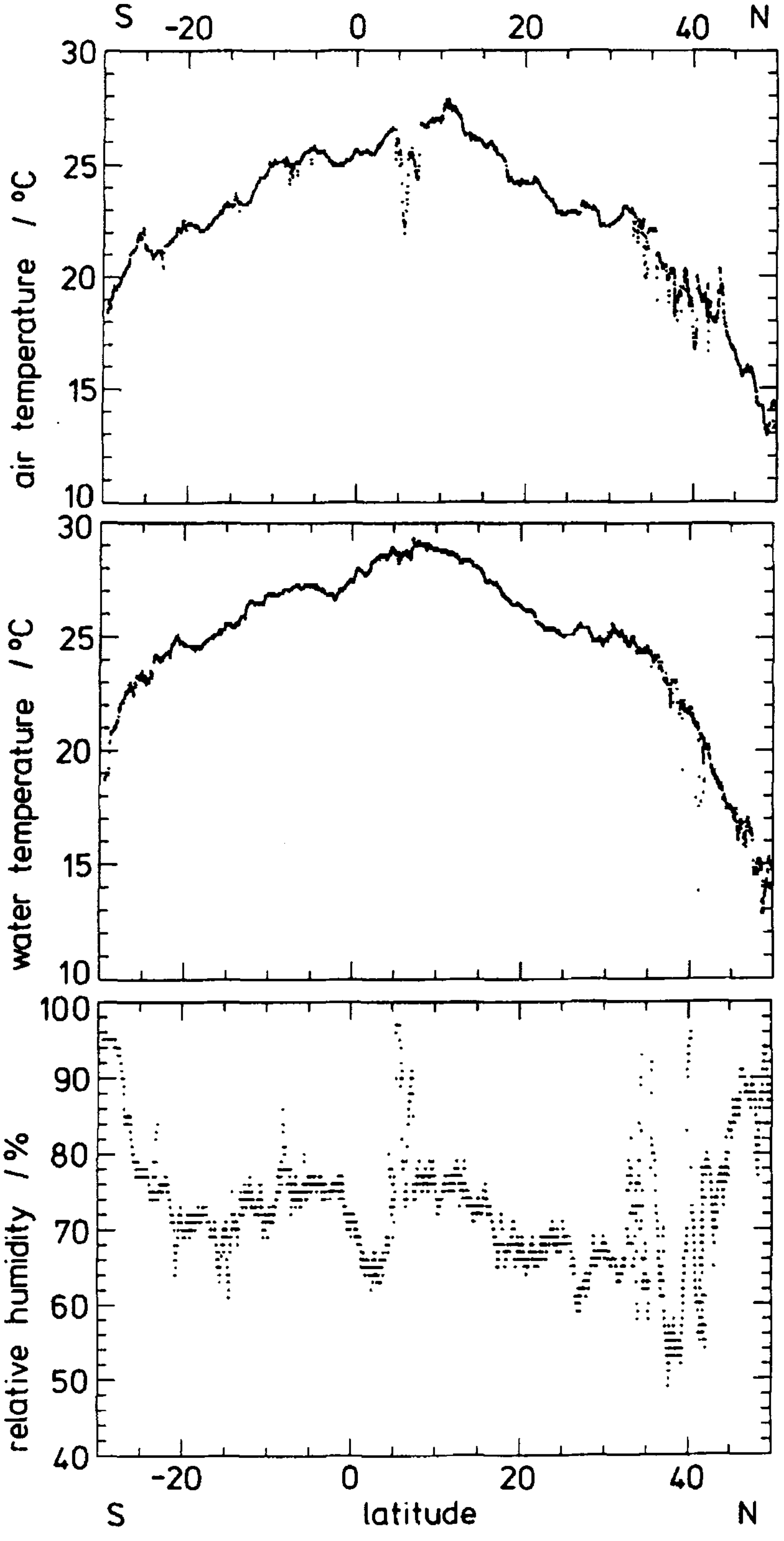

Fig. 3. True wind direction and speed, relative wind direction (counting bow as zero) and speed as a function of latitude during cruise ANT VII/1. Each point corresponds to a 10 min average.

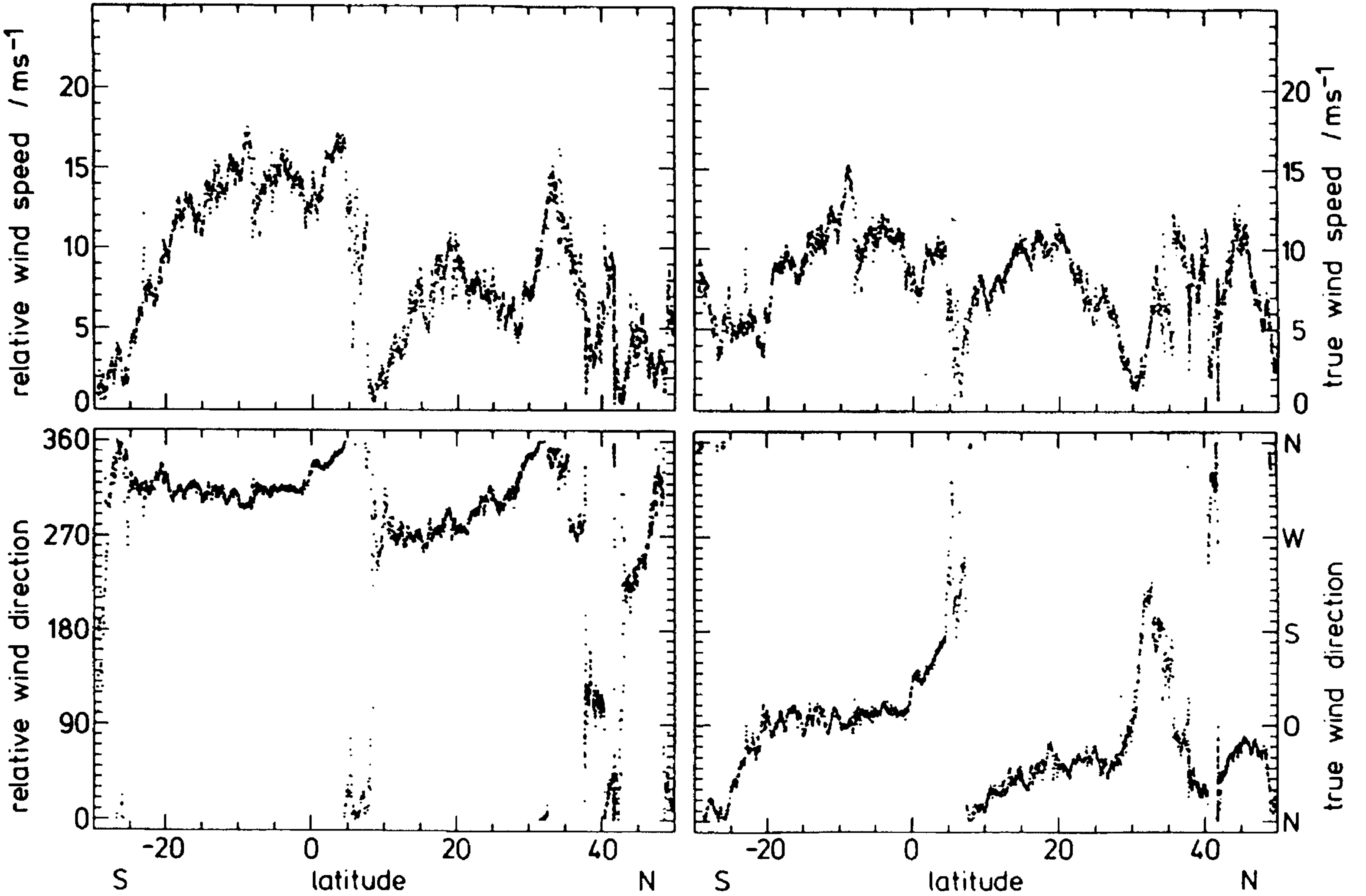

Fig. 4. Air and water temperature, and relative humidity as a function of latitude during cruise ANT VII/1.

4. Characterisation of Air Masses Encountered

For a detailed discussion of the individual experiments we refer to the papers describing the experiments cited in Table I and appearing in this issue. Here an overview of the measurements will be given by describing the various types of airmass encountered during the cruise (Table II).

From the start of the trip at Bremerhaven (54° N, 8° E), through the English channel, and south to about 40° N, air masses affected by industrial pollution prevailed. As summarized in Table II, this is indicated by elevated levels of acetylene (Koppmann *et al.*, 1990), condensation nuclei, ozone (Callies *et al.*, 1990; Brauers and Hofzumahaus, 1992 – this issue), and NO_x (Rohrer and Brüning; Harris *et al.*, 1992 – this issue). Also PAN mixing ratios up to 2 ppb were found, while this species remained undetectable (<0.4 ppt) for the rest of the cruise (Müller and Rudolph, 1992 – this issue). From 35° N to 15° N the atmospheric trace gas composition suggested that the airmass could be described as North Atlantic background. At tropical latitudes from 15° N to the ITCZ, which was centred around 5.5° N at the time of the cruise, continentally influenced air was indicated by elevated concentrations in the aerosol samples of crustal elements such as aluminium (Losno *et al.*, 1992 – this issue). The back trajectories support the interpretation that the sampled air masses had passed over the savannah and desert areas of North Africa. The levels of HNO_3 (Papenbrock *et al.*, 1992 – this issue), NO_x, C_2H_2, and CN, however, remained low, indicating that there was no major contribution from urban and industrial sources or from biomass burning.

In contrast, the airmasses encountered from south of the ITCZ to about 15° S (Table II) showed increased HNO_3 mixing ratios and higher C_2H_2, CO, O_3, and H_2O_2 than observed to the north and south of this latitude belt. CN levels were about 20 cm^{-3} immediately south of the ITCZ and south of 10° S with a local maximum of 100–300 cm^{-3} in between (Callies *et al.*, 1990); NO_x, however, remained at low levels. These observations fit the description of 'aged smog', with the long-lived combustion products such as CO and C_2H_2 being still present at elevated levels, and with higher levels of O_3 and H_2O_2 having been produced during the chemical evolution of the air mass, while the primary NO_x had been converted to HNO_3 during transport. The source of this disturbance was most likely biomass burning in equatorial Africa. This region of 'diluted and aged smog' coincides in season and location with the maxima of tropospheric ozone reported by Fishman *et al.* (1990).

Finally, south of 15° S until near the end of the measurements trace gas levels returned to maritime background levels. CN levels remained low until the last day when the high end of the range shown in Table II was encountered under the influence of the northerly winds resulting from the low pressure system at 40° S, 47° W.

Table II. Pollution levels and possible origin of the air masses encountered during ANT VII/1

Latitude	Type of airmass	CO[a]	HNO_3 ppt	NO_2 ppt	C_2H_2 ppt	CN cm^{-3}	O_3 ppb	Al $\mu g/m^3$	H_2O_2 ppb
50° N–35° N	NH polluted	90–120	na	na	100–260	10^3–10^5	30–58	0.3–1	0.0–1.5
35° N–15° N	N-Atlantic background	80–95	100–300	15–40	70–140	200–3000	28–32	<0.3	0.7–1.6
15° N–ITCZ	continental (Sahara)	80–90	0–80	60–90	40–120	20–3000	15–35	1.5–2.5	0.7–2.0
ITCZ–15° S	'aged' biomass-burning plume	80–150	0–270	10–60	60–160	20–300	28–36	<0.1	1.8–3.3
15° S–30° S	maritime background	70–90	0–70	10–50	40–70	20–1000	18–27	<0.1	1.0–2.4

ITCZ: Intertropical convergenze zone, location during the time of the cruise centered at 5.5° N.

[a] CO mixing ratios are relative data only (Koppmann *et al.* 1990).

5. General Conclusions from the Measurements

From a single cruise it is not possible to judge the frequency of such events as those experienced between 15° N and 15° S, or to ascertain their importance on the average on the global scale. However, the measurements carried out demonstrate that long range transport from the African continent can drastically perturb the conditions in the maritime boundary layer throughout very large regions of the Atlantic.

The 'background' conditions found between 30° and 15° in the Northern and between 15° and 25° in the Southern Hemisphere are likely more representative of the predominant state of the remote maritime boundary layer. Under these conditions, the diurnal variation in ozone, (Callies *et al.*, 1990; Brauers and Hofzumahaus, 1992 – this issue), the elevated H_2O_2 concentrations (Jacob and Klockow, 1992 – this issue) and the low NO and NO_2 levels (Rohrer and Brüning, 1992 – this issue; Harris *et al.*, 1992 – this issue) all indicate that ozone was being destroyed, rather than produced through photochemical activity in the boundary layer.

Atmospheric concentrations of several species were compared to sea-water mixing ratios, indicating that biologically inactive areas of the ocean were traversed by the ship. In particular, the short-lived organic sulphur species DMS and CS_2 (Pashalidis *et al.*, 1990; Staubes *et al.*, 1990) as well as a series of NMHC (Koppmann *et al.*, 1990; Plass *et al.*, 1992 – this issue) were investigated in that way. Even in these areas, the ocean is a source of light reactive hydrocarbons; this source is, however, not strong enough to have a major effect on the prevailing HCHO levels, (Harris *et al.*, 1992 – this issue; Carlier *et al.*, 1990).

The HCHO and gas-phase reactive hydrocarbon measurements may be used with a simple model to estimate the OH radical concentration (Harris *et al.*, 1992 – this issue) which according to this calculation lay in the range $3–6 \times 10^6$ cm^{-3} (mean noontime values) between 40° and 15° N. An second estimate of the OH concentrations may be obtained starting with the data of Hofzumahaus *et al.* (1992 – this issue) who report OH noontime production rates from the observed $J(O_3)$, $[O_3]$, and $[H_2O]$ in the range $4–8 \times 10^6$ cm^{-3} s^{-1}. By considering the levels of the main reaction partners for OH measured during the cruise, noontime OH concentrations in the range $4–7 \times 10^6$ cm^{-3}, are inferred. Despite the fact that the primary production of OH was high at tropical latitudes (due to high $J(O_3)$ and $[H_2O]$), the estimated OH concentrations were comparable to those observed (Platt *et al.*, 1988) or calculated (Perner *et al.*, 1987) for northern mid-latitude rural areas, because of the inefficient recycling at the very low NO levels encountered at low latitudes (Rohrer and Brüning, 1992 – this issue). A parallel consequence of the low NO should be a high HO_2 concentration and therefore a high H_2O_2 production via self-reaction of HO_2 radicals, in accord with the H_2O_2 mixing ratios measured under the background conditions (Jacob and Klockow, 1992 – this issue).

The measurements summarized here and presented in the companion papers constitute one of the few quite comprehensive data sets on the composition of the remote troposphere, which may be used to test present understanding of the oxidant chemistry of the maritime boundary layer.

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